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## Final Report for NAG-1-1224

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For the Global Troposphere Experiment project Pacific Exploratory Measurements West B (PEM West B), we made determinations of sulfur dioxide ( $\text{SO}_2$ ) and dimethyl sulfide (DMS) using gas chromatography - mass spectrometry with isotopically labelled internal standards. This technique provides measurements with precision of 1 part-per-trillion by volume below 20 pptv and 1% above 20 pptv. Measurement of DMS and  $\text{SO}_2$  were performed with a time cycle of 5-6 minutes with intermittent zero checks. The detection limits were about 1 pptv for  $\text{SO}_2$  and 2 pptv for DMS. Over 700 measurements of each compound were made in flight.

In contrast to the late summer period of Pacific Exploratory Mission-West A (PEM-West A) (1991) over this same area,  $\text{SO}_2$  showed little increase with altitude, and concentrations were much lower in the free troposphere than during the PEM-West B period. Volcanic impacts on the upper troposphere were again found as a result of deep convection in the tropics. Extensive emission of  $\text{SO}_2$  from the Pacific Rim land masses were primarily observed in the lower well-mixed part of the boundary layer but also in the upper part of the boundary layer. Analyses of the  $\text{SO}_2$  data with aerosol sulfate, beryllium-7, and lead-210 indicated that  $\text{SO}_2$  contributed to half or more of the observed total oxidized sulfur ( $\text{SO}_2$  plus aerosol sulfate) in free tropospheric air. The combined data set suggests that  $\text{SO}_2$  above 8.5 km is transported from the surface but with aerosol sulfate being removed more effectively than  $\text{SO}_2$ . Cloud processing and rain appeared to be responsible for lower  $\text{SO}_2$  levels between 3 and 8.5 km than above or below this region.

During both phases of PEM-West, dimethyl sulfide did not appear to be a major source of sulfur dioxide in the upper free troposphere over the western Pacific Ocean. In 1991 the sources of  $\text{SO}_2$  at high altitude appeared to be both anthropogenic and volcanic with an estimated 1% being solely from DMS. In 1994 the sources of  $\text{SO}_2$  were again dominated by the anthropogenic and volcanic emissions but with DMS estimated to be solely responsible for <10% of the  $\text{SO}_2$ . The primary difference for the increase in the DMS source was the very low concentration of  $\text{SO}_2$  at high altitude. In the midlatitude region near the Asian land masses, DMS in the mixed layer was lower than in the tropical region of the western Pacific.

Convective cloud systems near volcanoes in the tropical convergence in the western Pacific troposphere were a major source of  $\text{SO}_2$  at high altitudes during PEM-West B. High levels of  $\text{SO}_2$  were observed in several instances with large number concentrations of ultrafine CN above 9 km in the tropical convergence zone. Conversion of  $\text{SO}_2$  by OH to  $\text{SO}_3$  and subsequently to sulfuric acid may have been enhanced by lightning-produced NO levels exceeding 1 part per billion. Coupling of strong convection and volcanic sources of  $\text{SO}_2$  apparently is an important source of new particle formation at high altitude in the tropical convergence zone.

The results were reported in two publications in the PEM West B special issue of *Journal of Geophysical Research*.

Transport of sulfur dioxide from the Asian Pacific Rim to the North Pacific troposphere  
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